Magnetization Reversal in Ultrathin Ferromagnetic Films with Perpendicular Anisotropy: Domain Observations

J. Pommier, P. Meyer, G. Pénissard, and J. Ferré
Laboratoire de Physique des Solides, Bâtiment 510, Université Paris-Sud, 91405 Orsay, France

P. Bruno[(a)]
Institut d'Electronique Fondamentale, Bâtiment 220, Université Paris-Sud, 91405 Orsay, France

D. Renard
Institute d'Optique Théorique et Appliquée, Bâtiment 503, Université Paris-Sud, 91405 Orsay, France
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We report on the first observation of magnetic domain structure during spin reversal under field in ultrathin metallic films with perpendicular anisotropy. The magnetization reversal is dominated either by the nucleation process or by domain-wall motion. The related magnetic aftereffect is analyzed starting from the Fatuzzo theory.

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Ultrathin ferromagnetic metallic films or multilayers with perpendicular anisotropy are promising materials for magneto-optical (MO) recording.[1] Up to now, only little work has been devoted to the study of magnetization processes (coercivity, remanence, time-dependent effects, etc.) in these materials.[2,3] From a fundamental point of view, the question remains open whether the concepts elaborated for bulk materials on the existence of magnetic domains and walls can be extended to few monolayers (ML) thick metallic films or whether specific behaviors are to be expected.

The interpretation of striking dynamical aftereffects at room temperature, first seen on hysteresis loops,[3-5] requires a direct microscopic observation of the development of the magnetic domain structure at a constant magnetic field, lower than the coercive value H_c; this is an experimental challenge for ferromagnetic films of a few atomic layers with perpendicular anisotropy. The process of magnetization reversal is of considerable practical interest because of its role in thermomagnetic recording. The study of the mechanism of formation of domains, i.e., the nucleation of the reversed state and its growth, is required to determine the factors limiting the sensitivity in MO recording. For easy-plane-magnetized ultrathin ferromagnetic layers large domains have recently been observed in zero field.[6]

In this paper we focus on studies of the magnetization reversal under field of ultrathin hcp Co films of thicknesses $d < 6$ ML with perpendicular spin anisotropy, deposited on a Au(111) buffer layer. Since Au/Co/Au sandwiches also display square hysteresis loops[4,5] and giant magneto-optical effects[3] they are good candidates for MO storage media. We demonstrate that the magnetization reversal occurring just below the coercive field value involves a metastable phase where macroscopic spin "up" and spin "down" domains coexist. To analyze the spin reversal we have visualized the time dependence of the magnetic domain structure on a micrometer scale by means of a magneto-optic microscope.[7] The relaxation laws of the related magnetic aftereffect are then discussed in relation with the microscopic processes governing the dynamics of the magnetization. Among several studied samples we found two different typical magnetization reversal behaviors, even for films with comparable thicknesses. We have selected two samples to illustrate these two limiting cases. The preparation and structural properties of Au/Co/Au samples, grown on a glass substrate, have been described earlier.[8,9] Macroscopic MO measurements of the magnetization of Co films are performed by Faraday rotation on a 1-mm^2 area of the sample.[3] The microscopic magnetic imaging, realized by Faraday rotation, is improved by the use of a sensitive charge-coupled-device camera with high spatial resolution and subsequent image processing.[7]

Faraday-rotation hysteresis loops measured for samples I and II ($d = 5$ and 5.5 ML, respectively), for two field sweep rates, are shown in Fig. 1 after subtraction of the diamagnetic term.[3] The sample I displays a dynamic behavior similar to that observed in previously studied samples.[4,5] The apparent coercive field is time dependent; it decreases with lower sweep rate of the applied magnetic field.[4,5] The sample II exhibits a squarer hysteresis loop than sample I: The nucleation field H_N, at which the magnetization reversal (point S_N) starts, is better defined for sample II. The magnetic aftereffect in both samples is clearly related to thermally activated processes, as confirmed by the lack of time dependence of magnetic hysteresis loops at low temperature.

In accord with calculations[10] which predict very large domains for our samples (> 1 cm) we have observed no magnetic domain pattern in zero field for virgin films. The samples are first magnetized to saturation ($-M_S$) in a premagnetizing field $H_P = -1$ kOe applied perpen-

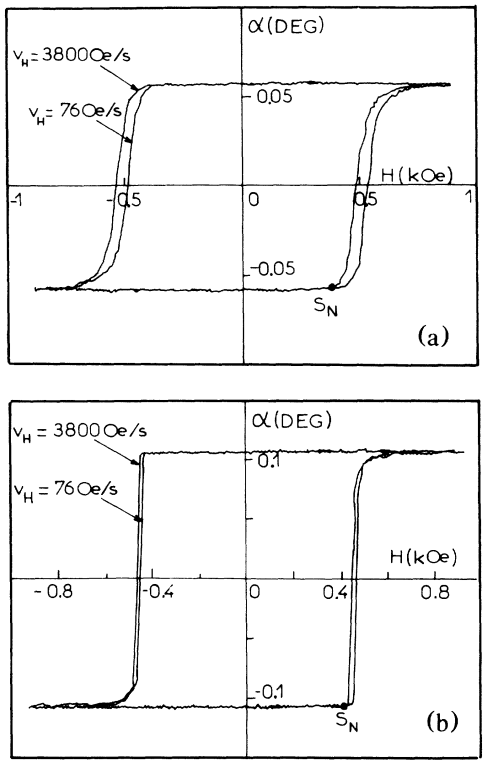

FIG. 1. Room-temperature Faraday-rotation hysteresis loops for (a) sample I and (b) sample II for two field sweep rates. The loops at 3800 Oe/s are averaged 32 times and the diamagnetic contribution subtracted.

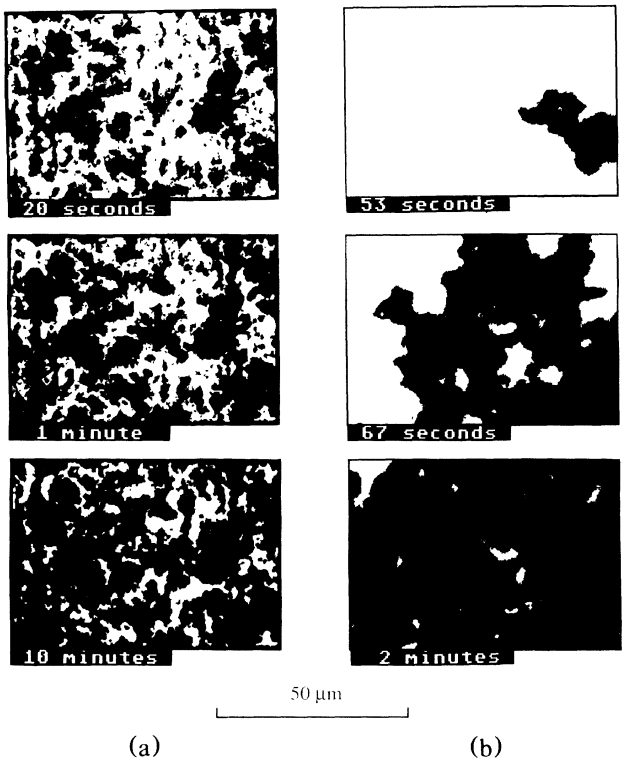

FIG. 2. Time evolution of the magnetic domain structure for (a) sample I at $H=470$ Oe and (b) sample II at $H=413$ Oe. "Spin up" domains (black) grow at the expense of the "spin down" state (white). The times, after application of H, are indicated in the pictures.

dicularly to the film plane. Then the field is reversed suddenly ($\tau_H=70$ ms) at $t=0$ to a positive value $H<H_c$. We demonstrate that all successive in-field magnetic domain patterns may be frozen by switching the field off, in agreement with the predominance of irreversible processes for magnetization ($M_R/M_S>0.99$ for $d=5$ ML).[4] This fact allows us to accumulate images of the same state and improve considerably the contrast of magnetic origin. Figure 2 gives the time evolution of the magnetic domain structure at room temperature for the two samples in fields small enough to ensure very slow activated dynamics. Reversed "spin up" domains develop in the initially created "spin down" state. The domains with magnetization M_S and $-M_S$, respectively, are separated by Bloch walls. The magnetization reversal may be analyzed considering the following different stages: (i) the nucleation of reversed domains, (ii) their local expansion from the nuclei, (iii) their widening by large domain-wall displacement, and (iv) the final reversal of small hard magnetic entities near crystal defects.

Dynamical processes can be analyzed from the relaxation of the macroscopic magnetization at constant field, close to its coercive value,[4,5] following the above premagnetizing procedure. In the present work the subsequent change in magnetization is recorded from the variation of the Faraday rotation (Fig. 3). Thus we can associate a particular domain pattern with any point of experimental relaxation curves.

For sample I, a nucleation-dominated reversal is clearly observed [Fig. 2(a)]. The starting of the spin reversal is dominated by the first two processes (i) and (ii): Nucleation centers appear randomly in the sample and domains grow irregularly from these nuclei over short length scales (<5 μm). Such films give rise to nonrectangular hysteresis loops [Fig. 1(a)] because of the local variations of the nucleation and propagation fields which provide a distribution of coercive fields through the sample. A similar behavior occurs in some amorphous Tb-Fe sputtered films.[11] This microscopic behavior explains well the magnetic aftereffect in sample I [Fig. 3(a)] and supports the previous interpretation that magnetization reversal occurs via thermally activated domain-wall motion involving short Barkhausen length.[4,5] Nucleation phenomena, however, were not discussed in Refs. 4 and 5.

For sample II a wall-motion-dominated reversal is evidenced at room temperature in Fig. 2(b). Below the coercive field the nucleation is a rare event; it is followed by the subsequent growth of domains by large wall

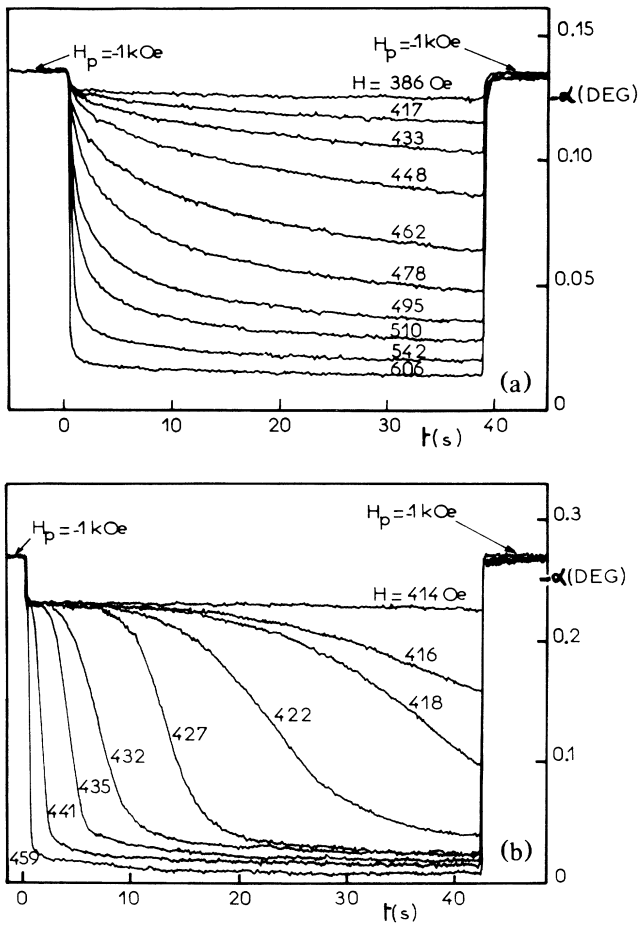

FIG. 3. Relaxation of the Faraday rotation for several values of the field for (a) sample I and (b) sample II.

motion [stages (i) and (iii)]. A small increase of H, promoting more nuclei, considerably accelerates the domain growth, explaining the relaxation data. In an early stage, the magnetization reversal comes from the development of a few reversed domains, widely separated from each other. The location of nucleation sites depends upon the magnetic history of the sample: We checked that the growth process is related to domain-wall propagation rather than nucleation of small adjacent domains existing near domain walls. A direct observation shows that magnetization develops through successive wall jumps of a few microns in amplitude, typical of a magnetic creep phenomenon. The wall surface is very ragged because of the local variations in coercivity due to defects and crystal roughness. In this case the propagation field H_P is undoubtedly smaller than the nucleation field H_N. For $H > H_N$, the domain walls expand rapidly through the film; that gives rise to a square hysteresis loop which does not depend much on the field sweep rate. From these microscopic observations it is straightforward to explain the magnetic aftereffect for sample II [Fig. 3(b)].

The magnetic relaxation rate is very sensitive to the applied field value H; it increases for both samples rapidly with H up to the saturation field H_S, for which the sample transforms in less than 5 ms into a single domain. More generally, at room temperature, the magnetization in ultrathin Co films can be reversed at a constant field $H < H_N$ by thermal activation after waiting long enough. This points out the metastable nature of the domain state. At low temperature, the spin down phase is stable up to the field $H = H_N$, for which an "instantaneous" reversal (less than 1 ms) of the largest part of the magnetization occurs by means of a few large Barkhausen jumps. As revealed by microscopic domain observations for both samples (Fig. 2) and by their relaxation magnetization curves (Fig. 3) the stage (iv) is always present in the final stage of the magnetization reversal. Local defects can modify the magnetic anisotropy thus generating hard magnetic centers which pin the initial magnetic state. The wall propagation stops temporarily at their boundary, as observed at long times in Fig. 2; it results in a lacunar domain structure which evolves slowly towards the uniform reversed state. The number of hard magnetic centers remaining at a given time is reduced when increasing the field H, reaching zero for $H = H_S$. The estimation of H_S is difficult since a few submicronic centers can always exist in the so-called saturated state. They act as natural nucleation centers for initiating domain growth by coming back to a reversed field. It is important to note that these two types of magnetization reversal mechanisms have been also observed in thick (0.1–1 μm) amorphous films of Tb-Fe,[12,13] for which usual magnetostatic considerations may be applied.

A theoretical model first developed by Fatuzzo[14] for polarization reversal in ferroelectric crystals, including both the nucleation process and domain-wall propagation, has been extended to interpret the magnetization reversal in GdTbFe thick films.[15] The form of the relaxation law strongly depends upon a parameter k proportional to the ratio of the domain-wall velocity v to the rate of nucleation R. Nucleation- and domain-wall-dominated reversals are associated to small (sample I) and large (sample II) k values, respectively. Calculated relaxation curves are in close agreement with experimental results (Fig. 3). As for GdTbFe films,[15] we checked, for each Co film at room temperature, that k is rather insensitive to H since all data fit a universal relaxation curve which scales with $t/\tau_{1/2}$. If H_0 stands for the coercive field [H_0(sample I) = 550 Oe, H_0(sample II) = 477 Oe] the characteristic time $\tau_{1/2}$, for which $M = 0$, is found to vary as

$$\tau_{1/2} = \exp A(H_0 - H) . \quad (1)$$

Such an expression (1) is expected for Bloch wall movements with $A = V_P M_S / k_B T$, V_P being the Barkhausen volume. The development of the magnetic domain structure (Fig. 2) suggests that both the nu-

cleation rate and domain-wall velocity (for $H < H_0$) are thermally activated and strongly depend on H in the following manner:[15]

$$R = R_0 \exp[-(E_N - 2HM_S V_N)/k_B T],$$
$$v = v_0 \exp[-(E_P - 2HM_S V_P)/k_B T]. \quad (2)$$

As found earlier,[4,5] the Barkhausen volume involved in the reversal of type-I samples is of the order of 10^{-7} μm^3, while for sample II, the detectable wall jumps allow us to estimate $V_P \sim 10^{-2}$ μm^3; these values are consistent with those of k. A severe limitation to the application of the Fatuzzo theory[14] comes from the assumption of the existence of only two activation energies E_N and E_P for nucleation and wall propagation mechanisms. The shape of the hysteresis loops for type-I samples may be interpreted only by assuming a Lorentzian distribution of activation energies.[5] A distribution of E_P is also expected for sample II since only a partial magnetization reversal occurs at H_N, as confirmed at low temperature. This means that, even for $k = 0$, the predicted exponential relaxation[14] cannot be observed. The relaxation curves for low-k values are better fitted by the universal law $M(t) = F[\ln(t/\tau_{1/2})]$, valid for activated dynamics associated with a distribution of energy barriers; we deduce $E_P/k_B T = 7.5$ for sample I. For sample II, even if in a first stage large domain-wall movements occur (large k value), hard magnetic sites prevent a final rapid reversal. This leads to a more localized spin reversal involving a smaller effective k value at long time.

Differences in sample preparation conditions were invoked to explain the drastic change in magnetization reversal observed in Tb-Fe amorphous thick films.[12,13] Since the two Co films were prepared in quasisimilar conditions the origin of such a behavior still remains unclear. Stress-induced effects, due in particular to the Au-Co lattice mismatch and sticking of the film on a glass substrate, may have strong effects on the nature of metastable states involved during spin reversal. A strong influence of roughness can be also emphasized since surface effects play a major role on the stability of the domain configuration.

In conclusion, we evidenced two limiting behaviors for magnetization reversal: nucleation- or wall-motion-dominated processes, but all intermediate situations were also observed. The dynamics of magnetization are undoubtedly strongly affected by weak changes in local properties which generate distributions of nucleation and propagation fields inside the Co film. Magnetic relaxation measurements and domain visualization at variable temperature are in progress to refine the dynamic analysis of spin reversal.

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[a]Presently on leave at the Institut für Angewandte Physik, Universität Regensburg, Regensburg, Federal Republic of Germany.

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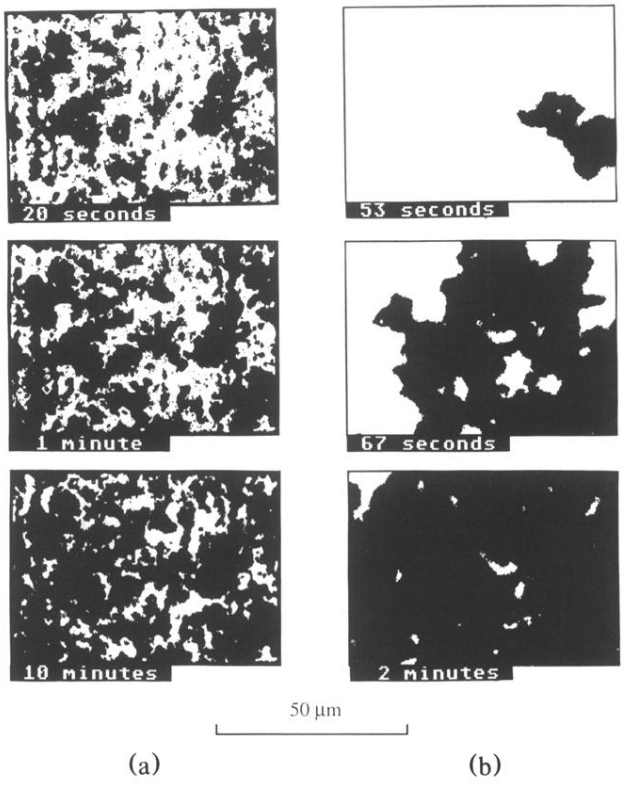

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